

REMARKS

Entry of the foregoing amendments and favorable consideration of the subject application is respectfully requested in view of the following comments.

The specification and claims have been amended to correct minor typographical errors and to make them more easily readable upon publication.

Specifically, in the textual description of the formulas $Ti(IV)O_aNbF_c$ and $MeTi(IV)O_aNbF_c$ identifying the nature of the subscripts a, b and c, the original specification had these reference letters, subscripted in the formulas and subscripted in the textual explanation which rendered it difficult to adequately discern these letters and the textual explanation somewhat difficult to read.

By the foregoing amendments, Applicants have corrected this formatting in the paragraphs presented herein and in the corresponding claims and respectfully request entry of these amendments in the subject application.

Applicants respectfully submit that the foregoing amendments are made to correct the typographical issues identified and that the subject application is in condition for allowance.

Applicants herewith submit substitute pages 3 and 4 of the specification, and the claims, pages 16 and 17, as amended and respectfully request that these pages be used in publication of the subject application.

An early notice of allowance is respectfully requested.

Respectfully submitted,

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acts by visible light. And the inventors of the present invention have continued intensive study to find out a compound which is stable under the reaction condition among these photo-catalysts, and found out that the compound composing of oxynitride containing at least one transition element can fulfill the function of the photo-catalyst which satisfy the above mentioned condition, and already proposed as the invention which dissolved said problem (JP Application 2000-256681; filed on August 28, 2000). Mostly of these compounds have perovskite-type crystalline structure, and the stabilizing effect under said photo-catalyst reaction condition is conjectured to be caused by this structural feature.

As a visible light active compound which was found out based on above mentioned conjecture, although a stable compound can be obtained among the compounds containing Ta or Nb, however, it was difficult to obtain a stable compound among the compound containing Ti(IV). Therefore, the inventors of the present invention have investigated the method how to obtain a useful compound as the visible light active photo-catalyst based on the theory of above mentioned hybridized oxynitride bonding orbital. In above mentioned consideration, the confirmation of characteristics based on above mentioned theory is considered to be useful.

The subject of the present invention is to provide a compound which is stable as a visible light active photo-catalyst having nitride bond of Ti(IV), further the object of the present invention is to provide a method for preparation of said compound. During the various considerations how to introduce a nitride bond into the compound containing Ti(IV), which has photo-catalytic activity, the inventors of the present invention found out that the introduction of nitride bond of Ti(IV) is possible when Ti(IV) contains F bond, and found out the synthesis of the compound containing Ti(IV) which has nitride bond by using compounds of TiO_aNbF_c or $MeTiO_aNbF_c$. And found that the obtained compound has a possibility to be a catalyst which is active by visible light, especially to be a catalyst which generate hydrogen or oxygen by photo splitting of water, thus the subject of the present can be accomplished. In the compounds of TiO_aNbF_c or $MeTiO_aNbF_c$, Me is an alkali earth metal such as Sr, c is 0.1 to 1, b is 0.1 to 1, desirably $b \geq 0.3$, and a is a value to be decided in relation to b and c.

By the way, when titanium oxide is nitrided with ammonia by the

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conventional method, Ti^{3+} is generated by reducing reaction, and when nitride reaction is moderated in order to suppress the reducing reaction, it becomes difficult to introduce sufficient nitrogen into material and the synthesis of the compound containing $Ti(IV)$ which has nitride bond is impossible, and the responsibility to visible light of the material is too low to absorb the visible light of around 600nm. Accordingly, the method for synthesis of the compound containing $Ti(IV)$ which has nitride bond is an epoch-making invention.

SUMMARY OF THE INVENTION

The first one of the present invention is a photo-catalyst containing titanium fluoride nitride comprising, $Ti(IV)O_aNbF_c$ or a compound represented by $MeTi(IV)O_aNbF_c$ prepared by doping at least one metal Me selected from the group consisting of alkalis or alkali metals on $Ti(IV)O_aNbF_c$ (wherein, b is 0.1 to 1, c is 0.1 to 1 and a is a value to maintain $Ti(IV)$ and is decided in relation to b and c.). Desirably, the present invention is the photo-catalyst containing titanium fluoride nitride, wherein $Ti(IV)O_aNbF_c$ possesses anataze structure and $MeTi(IV)O_aNbF_c$ possesses perovskite to anataze structure. Further desirably the present invention is the photo-catalyst containing titanium fluoride nitride to which at least one promoter selected from the group consisting of Pt, Ni and Pd is loaded.

The second one of the present invention is a photo-catalyst for water splitting containing titanium fluoride nitride comprising $Ti(IV)O_aNbF_c$ or a compound represented by $MeTi(IV)O_aNbF_c$ prepared by doping at least one metal Me selected from the group consisting of alkalis or alkali metals on $Ti(IV)O_aNbF_c$ (wherein, b is 0.1 to 1, c is 0.1 to 1 and a is a value to maintain $Ti(IV)$ and is decided in relation with b and c.). Desirably, the second one of the present invention is a photo-catalyst for water splitting containing titanium fluoride nitride wherein $Ti(IV)O_aNbF_c$ possesses anataze structure and $MeTi(IV)O_aNbF_c$ possesses perovskite to anataze structure. Further desirably the second one of the present invention is a photo-catalyst for water splitting containing titanium fluoride nitride to which at least one promoter selected from the group consisting of Pt, Ni and Pd is loaded.

The third one of the present invention is a method for preparation of a

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CLAIMS

1. A photo-catalyst containing titanium fluoride nitride comprising, $Ti(IV)O_aNbF_c$ or a compound represented by $MeTi(IV)O_aNbF_c$ prepared by doping at least one metal Me selected from the group consisting of alkali or alkaline earth metals on $Ti(IV)O_aNbF_c$, wherein, b is 0.1 to 1, c is 0.1 to 1 and a is a value to maintain $Ti(IV)$ and is decided in relation to b and c.
2. The photo-catalyst containing titanium fluoride nitride of claim 1 to which at least one promoter selected from the group consisting of Pt, Ni and Pd is loaded.
3. The photo-catalyst containing titanium fluoride nitride of claim 1, wherein $Ti(IV)O_aNbF_c$ possesses anatase structure and $MeTi(IV)O_aNbF_c$ possesses perovskite to anatase structure.
4. The photo-catalyst containing titanium fluoride nitride of claim 3 to which at least one promoter selected from the group consisting of Pt, Ni and Pd is loaded.
5. A photo-catalyst for water splitting containing titanium fluoride nitride comprising, $Ti(IV)O_aNbF_c$ or a compound represented by $MeTi(IV)O_aNbF_c$ prepared by doping at least one metal Me selected from the group consisting of alkali or alkaline earth metals on $Ti(IV)O_aNbF_c$, wherein, b is 0.1 to 1, c is 0.1 to 1 and a is a value to maintain $Ti(IV)$ and is decided in relation with b and c.
6. The photo-catalyst for water splitting containing titanium fluoride nitride of claim 5 to which at least one promoter selected from the group consisting of Pt, Ni, Ru and Pd is loaded.
7. The photo-catalyst for water splitting containing titanium fluoride nitride of claim 5, wherein $Ti(IV)O_aNbF_c$ possesses anatase structure and $MeTi(IV)O_aNbF_c$ possesses perovskite to anatase structure.

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8. The photo-catalyst for water splitting containing titanium fluoride nitride of claim 7 to which at least one promoter selected from the group consisting of Pt, Ni and Pd is loaded.

9. A method for preparation of a photo-catalyst represented by $Ti(IV)O_aNbF_c$, wherein a, b and c are same as to claim 1, by baking titanium di-ammonium fluoride halide represented by $(HH_4)_2TiF_dX_{6-d}$, wherein, d is integer or 1-6, which contains at least F and ammonium halide by the ratio of equimolar or by the ratio of slightly excess of ammonium halide at the maximum temperature from 200°C to 500°C so as to form a starting material, then said starting material is nitrogenated by thermal synthesis in ammonia atmosphere containing from 0.02% to 10.00% of oxygen, air or water to ammonia by reduced mass to oxygen atom at the maximum temperature from 350°C to 700°C for over than 5 hours.

10. A method for preparation of a photo-catalyst represented by $SrTi(IV)O_aNbF_c$, wherein a, b and c are same as to claim 1, by baking titanium di-ammonium fluoride halide represented by TiF_dX_{6-d} and/or $(HH_4)_2TiF_dX_{6-d}$, wherein x and d are integer of 1-6, which contains at least F and at least one compound selected from the group consisting of SrO, SrOH and SrX so as to form a starting material or $SrTiF_6$, then said starting material or $SrTiF_6$ is nitrogenated by thermal synthesis in ammonia atmosphere containing from 0.02% to 10.00% of oxygen, air or water to ammonia by reduced mass to oxygen atom at the maximum temperature from 350°C to 700°C for over than 5 hours.